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Materials at Atomic Pressure

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Materials at atomic pressures

NIF facility time proposal; PI: Damien Hicks
Platform: Materials research, Streaked radiography, using an ignition-type target and pulse

Scientific discussion

Atomic units give the scale at which quantum processes operate. As combinations of fundamental constants they concisely encapsulate qualities of the atom, e.g. atomic length and energy scales are given by the Bohr radius and the Hartree (or Rydberg) respectively. Although many of these quantities were probed in the early part of the 20^{th} Century the atomic unit of time, at tens of attoseconds, was first probed in 2001. Today, the only atomic unit that remains to be studied experimentally is the atomic unit of pressure, at 294 Mbar (or 147 Mbar in Rydberg atomic units). This is nature's definition of the 'high' in high-pressure science, and it sets the scale for new physics and chemistry. Among experimental facilities, only the NIF can attain and accurately probe atomic pressures. We propose to directly study material properties at these conditions by examining the short-range ordering of atoms using x-ray absorption fine structure spectroscopy (XAFS) of layers in spherical, ignition-type imploding shells.

What happens at atomic pressures? The atomic unit of pressure represents the quantum mechanical pressure exerted by an orbiting electron to prevent collapse into the nucleus. Applying external pressure of this magnitude seriously disrupts orbitals and alters the character of the atom itself. Core electron orbitals overlap and chemical bonds are no longer constrained to occur between valence electron orbitals alone. The most direct probe of bonding requires a short-range order diagnostic such as XAFS.

Proposed method

The proposed research will build upon (i) Many of the same capabilities already developed for the ignition program (symmetry, pulse-shaping, pre-heat control, and target fabrication) in order to prepare a homogeneous atomic-pressure state in a compressed spherical shell, and (ii) Recent Mbar XAFS measurements recently performed by this team on planar samples at OMEGA. Performing XAFS in a spherical implosion is new and, due to the stringent requirements of high symmetry and careful adiabat shaping, can only be performed on the NIF.

Standard NIF ignition pulses create drive pressures of ~100 Mbar in an imploding shell, rising to tens of Gbar upon stagnation (see Fig. 1) . The test sample (Ge or Fe) would be sandwiched inside a CH (or diamond) ablator, in the same way that Ge-doped plastic is in the current ignition design. Design simulations will be required to determine how to tune the drive pulse to avoid pre-heat of the iron and to place it in on a low adiabat. The in-flight pressure will be measured using the DISC or GXD, as has already been done on NIF, by determining the acceleration in the 1-D radiograph (see Fig. 2).

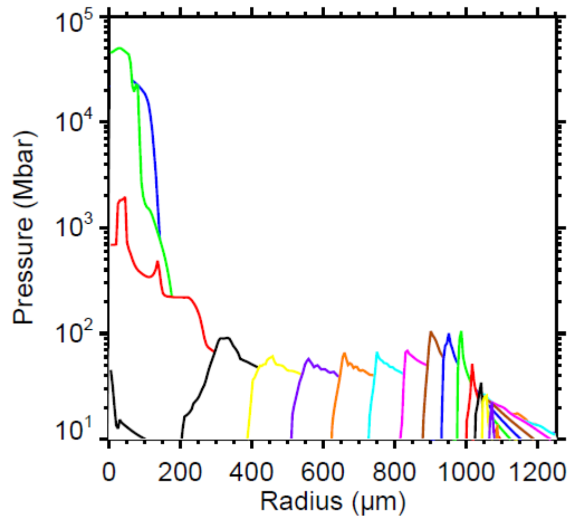

Figure 1: Convergence of a CH capsule indirectly-driven by a NIF ignition-type pulse. This shows how initial ablation pressures of around 100 Mbar reach 10's of Gbar upon convergence. Considerable effort has been invested by the ICF ignition program to achieve these conditions symmetrically. Our study will leverage such existing capabilities to prepare iron embedded in a spherical ablator on a low adiabat at atomic pressures.

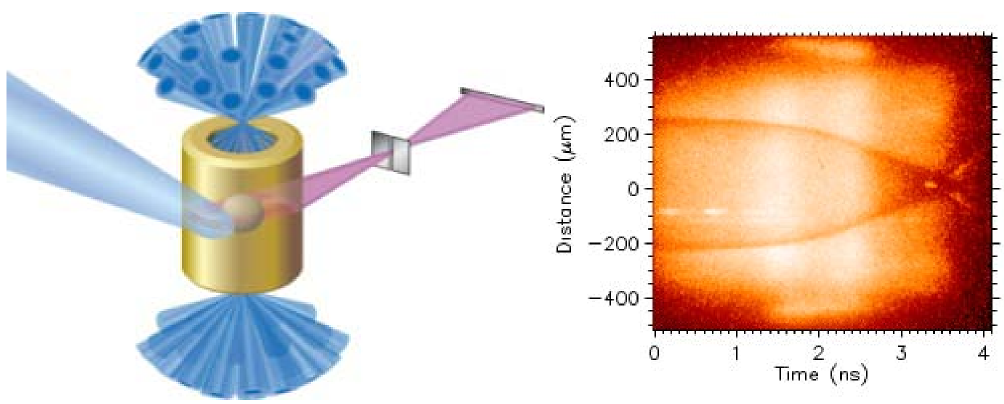

Figure 2: Streaked x-ray imaging of an imploding capsule will be used to determine the pressure and density profiles inside the imploding capsule via regularized inversion. This was recently achieved on NIF.

X-ray absorption fine structure (XAFS) will be used to determine the short range order, in particular the peak in the radial distribution function g(r), of the compressed material as well as (via XANES) more detailed bonding characteristics. This method relies on the scattered photoelectron wave modulating the x-ray absorption above the K-edge (or any other edge). Very recently XAFS of multi-shock compressed iron at 3 Mbar was demonstrated by this group – several times higher pressure than has ever been achieved before (Fig 3, 4). This was done using a new technique whereby a thin iron sample was tamped between two thin diamond plates. On NIF, the spherical analogue of this is to bury the sample layer inside the shell of a spherical capsule. The bright flash of x rays produced at the center of the capsule upon stagnation of the fill gas provides the broadband source of x rays. A flat crystal spectrometer (HSXRS) will be used to determine the absorption spectrum above the K-edge.

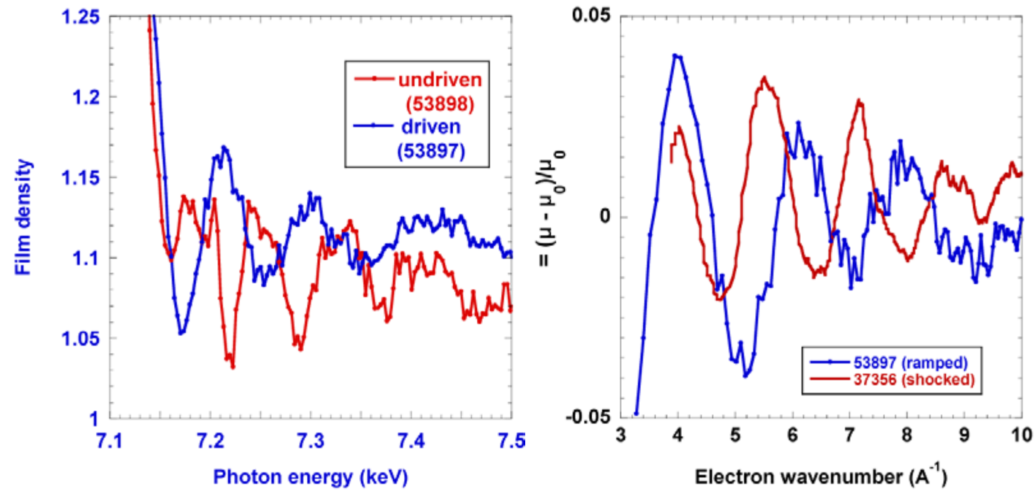

Figure 3: *Recent results at OMEGA have shown that it is feasible to trap a thin iron sample between diamonds dynamically loaded to several Megabar. This produces a spatially and temporally homogeneous state of compressed iron which is then probed using the broadband x ray spectrum. The effect of increasing compression is exhibited in the loss of the bcc peak (at 7.17 keV) after transition to the hcp phase and the significant increase in oscillation period. Adapting this to a spherically imploding shell much like existing ignition capsule designs provides access to 100+ Mbar states.*

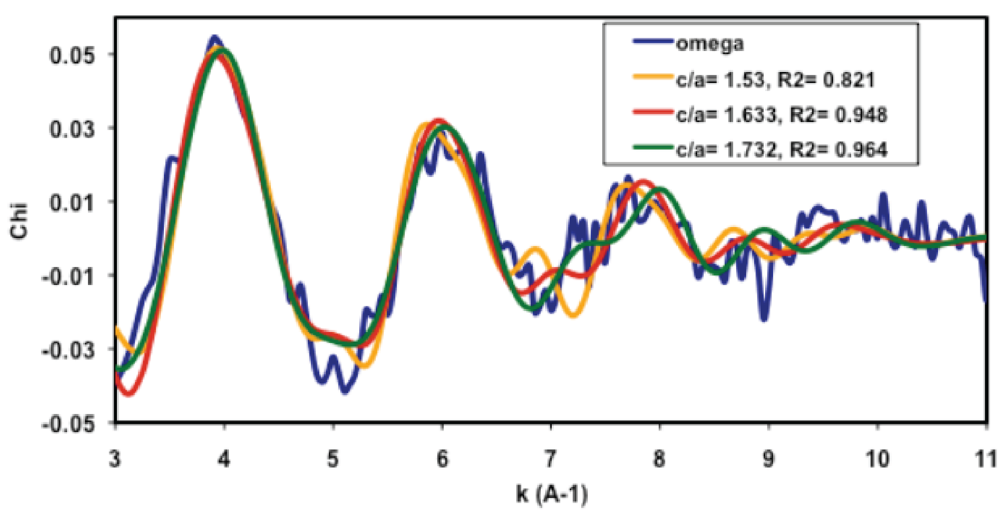

Figure 4: The FEFF8 scattering code has been used to fit the XAFS data at 3.5 Mbar. A best fit using multi-scattering calculations with up to 12 shells found that the crystal phase was hcp with ρ/ρ_0=1.83$\pm$0.13, T=5700$\pm$1300 K, and c/a=1.732. The bcc phase could be ruled out entirely while the fcc phase was slightly less preferable than hcp. The fact that calculations using 12 shells provided a better fit than using fewer shells strongly suggests that the material is still solid under these conditions, in line with calculations of the Fe melt line.

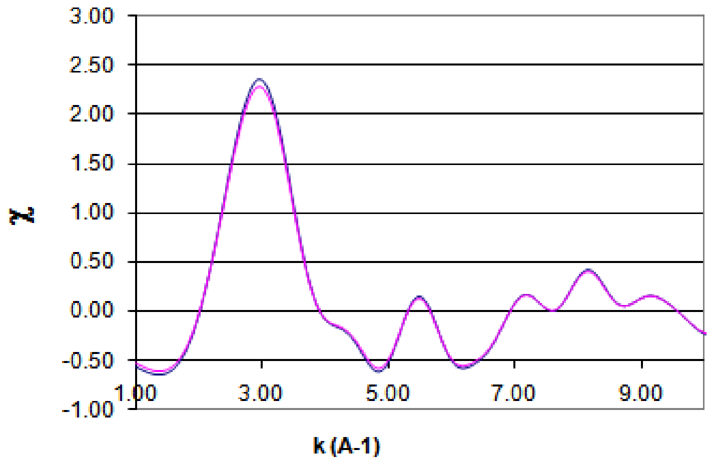

Figure 5: Predicted XAFS spectrum for Fe at 550 Mbar shows oscillation amplitudes far greater than have ever been observed before (compare the χ-scale in Figure 4 which only shows amplitudes of ± 0.05) . This is in part because modulations in the photoabsorption (χ) are proportional to $1/R^2$, where R is the distance between atoms. Compression to atomic pressures causes a dramatic increase in the XAFS signal to the extent that it is no longer "fine" structure.

Experimental feasibility

Several aspects of this experiment make it suited to NIF alone. Because of the severe requirements of spherical symmetry and high energy density the only other facility where this experiment might be performed is OMEGA. Pulse shaping and low M-band will be critical to keep the iron sample on a low adiabat at high pressure – ruling out OMEGA. Also, the large energies of NIF will be required to bring a sufficient volume of homogeneous sample up to high pressures and to hold it there for the 100+ ps required for the measurement.

Experimental team

This project will be undertaken by several members of the Planetary Sciences University Use of NIF Consortium led by Raymond Jeanloz of U.C. Berkeley, in particular: Damien Hicks, LLNL, hicks13@llnl.gov, 925-424-5220, Australia; Yuan Ping, LLNL, ping2@llnl.gov, China. This project is synergistic with concurrent proposals submitted by Peter Norreys (opacity) and Roger Falcone (ultra-high pressure).

Required capabilities and resources

Laser parameters: 1-1.3 MJ, close to NIF Scale 1.07 pulse shape.
Beam geometry: Standard ignition hohlraum + 2 quads for backlit implosion
Diagnostics: HSXRS@~7-12 keV, DISC (or GXD version)
Target fabrication of hohlraum, capsules (including special coatings)
Radiation hydrodynamic design simulations to define pulse shapes
Shot time: 2010 – 2 shots to test achievement of pressure and density states using DISC
2011 – 2 shots to explore pre-heat and adiabat control using absorption spectra
2012 – 3 shots to do fully integrated streaked imaging and absorption spectra

Instructions

The National Ignition Facility

1. This template is designed to gather basic facility information regarding experiments proposed under the FY2010 NIF facility time call.

2. The template is broken into 5 sections:
   a) Summary of proposed experiment: Desired platform (if known), NIF shots requested, brief campaign description, sketch of experimental configuration
   b) Diagnostic requirements
   c) Laser requirements
   d) Target requirements
   e) Other requirements

3. Please fill out each section and keep your answers brief. The NIF team will request additional information as needed from the Principal Investigators. Please attach additional pages to any section as needed.

4. Further information on the facility and the NIF call may be found at: https://lasers.llnl.gov/for_users/experimental_capabilities/

5. Thank you for your assistance, and please contact the NIF User Office if you have questions.

Summary of proposed experiment (Page 1 of 3)

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• Desired platform (If known): *(Fill in or indicate "not applicable")*
(Available platforms include: a) Capsule implosions; b) Hohlraum energetics;
c) Radiation transport; d) Shock timing; e) Streaked radiography; f) X-ray opacity; g)
X-ray sources. For further information on platforms see the NIF website:
https://lasers.llnl.gov/for_users/experimental_capabilities/index.php)
Streaked radiography

• Number of shots requested: Please fill out table below indicating number of "good data" shots requested each year. Do not add in additional shots to account for contingency, experimental problems, etc; NIF staff will consider this in planning evaluation

Summary Shot Table	*FY2010*	*FY2011*	*FY2012*	**Comments**
Total shots	*2*	*2*	*3*	*(Fill in if desired)*

Summary of proposed experiment (Page 2 of 3)

The National Ignition Facility

- **Brief campaign description (include summary of preparatory shots (drive, diagnostic development, other) and actual data acquisition shots):**

- **FY10 shots: Designed to be essentially identical to Scale544 ignition shot. Debug streaked radiography diagnostic to confirm measurement of velocity and ablation pressure. First measurement of Germanium K-edge.**

- **FY11 shots: Re-design pulse shape and target to minimize pre-heat and mix and thus sharpen XAFS signal.**

- **FY12 shots: Re-design target to use Fe instead of Ge. Measure XAFS.**

Summary of proposed experiment (Page 3 of 3)

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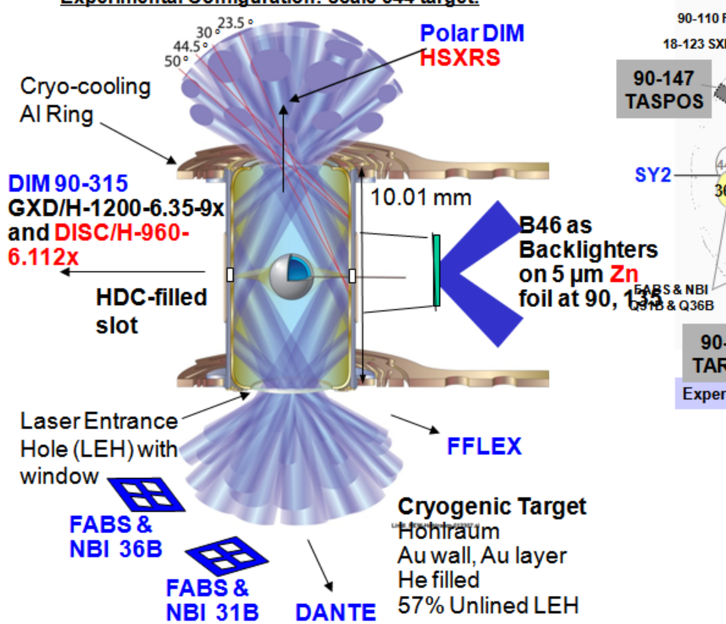
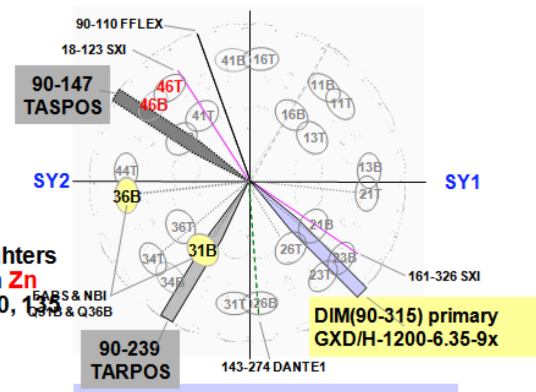
• **Sketch of experimental configuration: Pls. provide a simple sketch of the experimental configuration below. Include orientation of target, laser and any backlighter beams, diagnostic sightlines, etc. If configuration is identical to an existing platform so indicate. For further information on existing platforms and chamber geometry see the NIF website:**
https://lasers.llnl.gov/for_users/experimental_capabilities/index.php

Experimental Configuration: scale 544 target:





Experiment Layout - Target Chamber (Top View)

Diagnostic requirements

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• Please refer to the diagnostic list on NIF user website:
https://lasers.llnl.gov/for_users/experimental_capabilities/diagnostics.php

• List below NIF diagnostics required for your experiment (along with a short summary description of required spatial, temporal, and spectral resolution) *or* describe what you wish to observe, and NIF staff will match to available diagnostics.

- HSXRS from 7-13 keV, 20 eV resolution
- DISC w/ 4ω fidu - < 50 ps temporal, < 25 μm spatial

• Also indicate below if any additional, user provided diagnostics are required. Provide a short summary of the user provided diagnostic below, including a list of all materials to be introduced into the target chamber.

PI name Hicks, Damien   Proposal title: Materials at atomic pressures

Laser requirements (1 of 2)

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Laser Parameter	Value
1) Platform to be used	*Streaked radiography*
2) Number of beams required	*192*
3) 3ω energy desired per beam (Maximum allowed: 3 kJ (2nsec square); for pulses other than 2nsec square provide plot of desired power vs. time on next page. NIF User Office will inform users if energy requirements exceed allowable.)	*4-5 kJ/beam*
4) Peak power per beam (350 TW maximum total peak power for shaped, ignition-like pulses)	*1.5-2 TW/beam*
5) Pulse shape (up to 20 nsec duration) (Options: Square, impulse (88 psec), or shaped; provide plot of desired power vs. time for shaped pulse on next page)	*Scale544 ignition pulse*
6) SSD bandwidth (options- 45 to 90 GHz, 45 GHz default)	*45 GHz (modify if desired)*
7) Focal spot size (~250-μm (unconditioned) or ~1-mm (conditioned))	*Scale 1, 1.07 CPP*
9) Delays between beams (up to 10 nsec-all pulses in a quad must have same delay)	*Bkl beam delayed ~15 ns*
10) Backlighter beam energy, pulse duration	0.5 TW 2 ns FWHM Gaussian followed by 1.9 TW 1.5 ns FWHM Gaussian 4.3 ns later, chopped in front and back (ConvAbl style)
11) Other specifications	*Specify if desired*

Laser requirements (2 of 2)

For shaped pulses, sketch desired power vs. time below:

Standard ignition shaped pulse

Target requirements (1 page per target type)

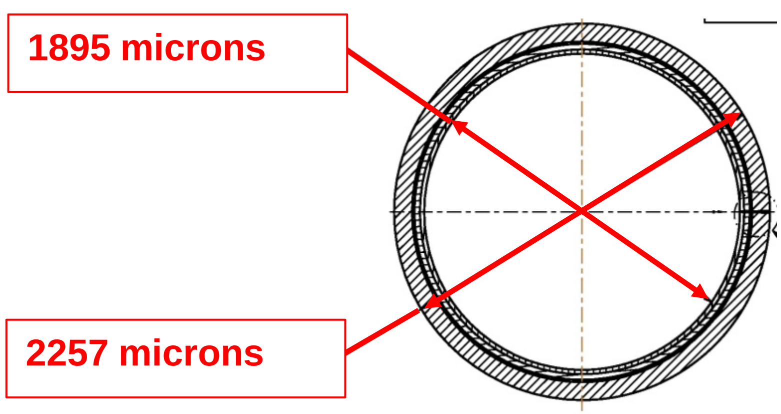
- List target types required (example: drive measurement; diagnostic test; data acquisition target)

- For each target type provide a sketch of the target below. Include dimensions and a list of *all* materials to be used. Also indicate any critical tolerances required, and indicate components (if any) to be provided by the Principal Investigator.

•See next 4 pages

Physics Requirements - Capsule

Rqmt Title	Rqmt Text	Basis Intent (at 20K)
CH Capsule	Inside diameter	1895 +/- 30 microns
	4 Layers (inside to outside)	1. 26 microns CH 2. 34 microns CH, Ge 0.6% 3. 14 microns CH, Ge 0.3% 4. 107 microns CH
	Wall thickness	181 +/- 5 microns



Physics Requirements – Hohlraum

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Rqmt Title	Rqmt Text	Basis Intent (at 20K)
Hohlraum	Hohlraum inside diameter	5.440 +/- 0.01 mm
	Hohlraum length	10.01 +/- 0.02 mm
	LEH opening diameter	3.101 +/- 0.01 mm
	AuB Hohlraum liner thickness	1.2 +/- 0.1 microns
	Pressurization	400 Torr
	Nominal hohlraum Gaps	Hohlraum halves - 20 microns Hohlraum half to LEH - 20 microns

Physics Requirements – Hohlraum

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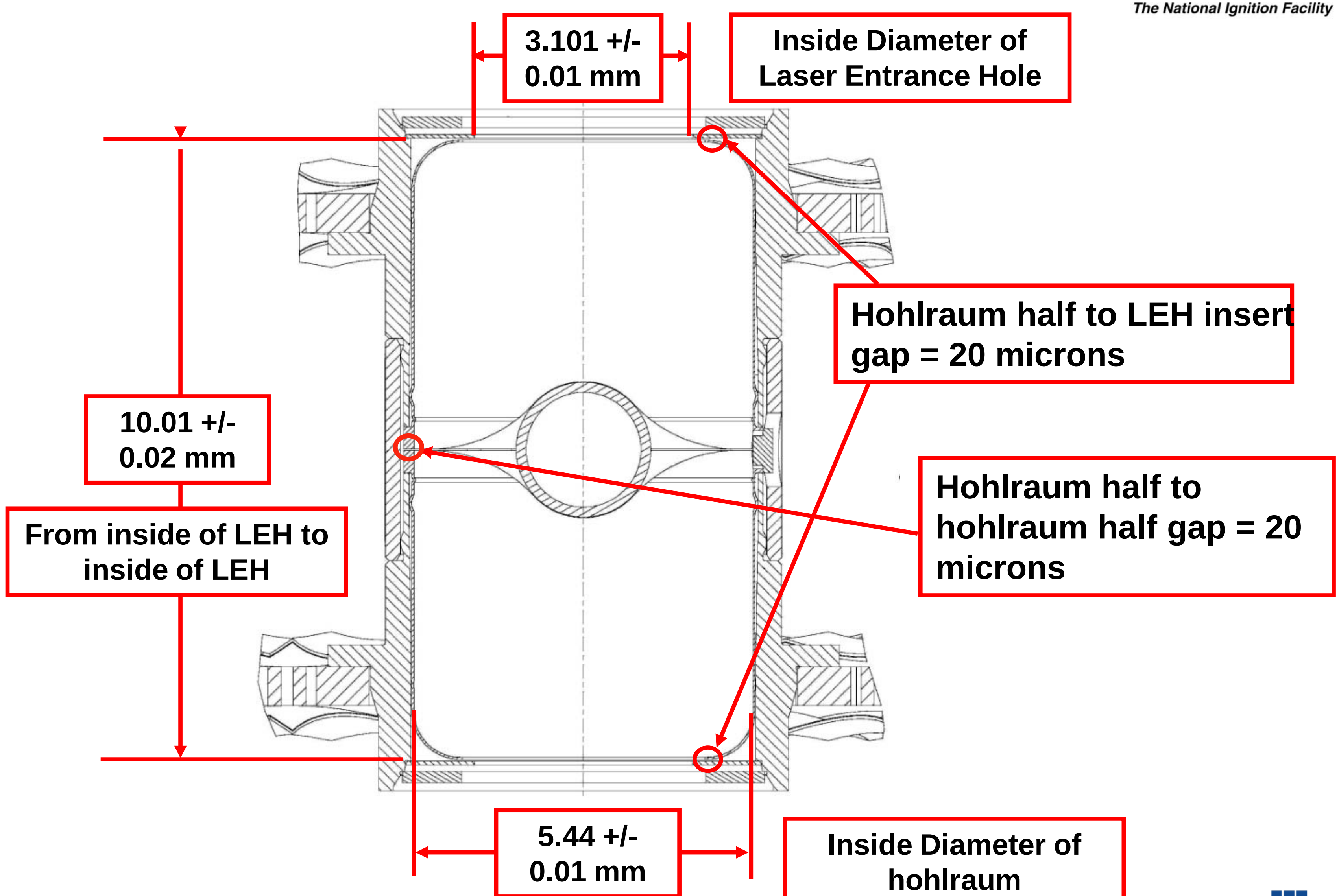

Physics Requirements – Diagnostic

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Rqmt Title	Rqmt Text	Basis Intent (at 20K)
Hohlraum HDC slots	Nominal width	1.2 mm
	Maximum one-side gap	0.02 mm
90-315 Diagnostic HDC Window	Height	0.146 +/- 0.002 mm
	Width	1.15 +/- 0.01 mm
	Radial Thickness	0.170 +/- 0.01 mm
90-135 Backlighter HDC Window	Height	0.102 +/- 0.002 mm
	Width	1.15 +/- 0.01 mm
	Radial Thickness	0.170 +/- 0.01 mm

- **HDC windows will match the outer radius of the hohlraum flange and be >= flush**
- **HDC windows can re-enter the hohlraum up to 60 microns**

Other requirements

The National Ignition Facility

- **Indicate other requirements (electrical, vacuum,…) for your proposed experiment. For any items to be introduced into the target chamber not mentioned to this point, please list *all* materials to be used.**